

LARGE EXCESS OF HEAVY NITROGEN IN BOTH HYDROGEN CYANIDE AND CYANOGEN FROM COMET 17P/HOLMES

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ABSTRACT

From millimeter and optical observations of the Jupiter-family comet 17P/Holmes performed soon after its huge outburst of October 24, 2007, we derive $^{14}\text{N}/^{15}\text{N}=139\pm 26$ in HCN, and $^{14}\text{N}/^{15}\text{N}=165\pm 40$ in CN, establishing that HCN has the same non-terrestrial isotopic composition as CN. The same conclusion is obtained for the long-period comet C/1995 O1 (Hale-Bopp) after a reanalysis of previously published measurements. These results are compatible with HCN being the prime parent of CN in cometary atmospheres. The ^{15}N excess relative to the Earth atmospheric value indicates that N-bearing volatiles in the solar nebula underwent important N isotopic fractionation at some stage of Solar System formation. HCN molecules never isotopically equilibrated with the main nitrogen reservoir in the solar nebula before being incorporated in Oort-cloud and Kuiper-belt comets. The $^{12}\text{C}/^{13}\text{C}$ ratios in HCN and CN are measured to be consistent with the terrestrial value.

Subject headings: comets: general — comets: individual (17P/Holmes) — radio lines : solar system

1. INTRODUCTION

Comets are made of ices, organics and minerals that record the chemistry of the outer regions of the primitive solar nebula where they agglomerated 4.6 Gyr ago. Compositional analyses of comets can provide important clues on the chemical and physical processes that occurred in the early phases of Solar System formation, and possibly in the natal molecular cloud that predated the formation of the solar nebula (Ehrenfreund et al. 2005). In particular, isotopic ratios in cometary volatiles are important diagnostics of how this matter formed, since isotopic fractionation is very sensitive to chemical and physical conditions. However, such measurements are rare. Strong deuterium enhancements are observed in H₂O and HCN gases of several comet comae that are characteristic of interstellar or protosolar chemistry at low temperature (Ehrenfreund et al. 2005; Bockelée-Morvan et al. 2005). On the other hand, questions arise of how to explain the non-terrestrial and uniform values of the $^{14}\text{N}/^{15}\text{N}$ isotopic ratio in CN (Arpigny et al. 2003; Hutsemékers et al. 2005; Manfroid et al. 2005; Jehin et al. 2004), also indicative of interstellar-like chemistry, while this ratio was measured to be terrestrial in HCN (Jewitt et al. 1997; Ziurys et al. 1999), the presumed source of CN radicals in cometary atmospheres. Here we present measurements of the $\text{HC}^{14}\text{N}/\text{HC}^{15}\text{N}$ and

$\text{H}^{12}\text{CN}/\text{H}^{13}\text{CN}$ isotopic ratios acquired in comet 17P/Holmes by millimeter spectroscopy, together with $\text{C}^{14}\text{N}/\text{C}^{15}\text{N}$ and $^{12}\text{CN}/^{13}\text{CN}$ measurements from visible spectroscopy. The brightness of this short-period (6.9 years) comet of the Jupiter family unexpectedly increased (from total visual magnitude $m_v = 17$ to $m_v = 2.5$) on October 24, 2007, while it was at a distance of 1.63 AU from the Earth and at 2.44 AU from the Sun (IAU Circ. 8886). This huge outburst of activity, which is likely related to a sudden fragmentation of the nucleus followed by the subsequent production of a large quantity of grains, offered us the opportunity to search for weak spectral signatures of rare isotopes using complementary techniques.

2. THE $^{14}\text{N}/^{15}\text{N}$ RATIO IN HCN IN 17P/HOLMES

We carried out observations of 17P/Holmes using the 30-m telescope of the Institut de Radioastronomie Millimétrique (IRAM) located in the Sierra Nevada (Spain). Isotopic measurements were performed on October 27–28 UT. The tracking of the comet was done using orbital elements K077/06 from JPL Horizons system. The pointing of the telescope was checked and updated by repeated observations of a nearby quasar. Sky cancellation was performed by wobbling the secondary mirror with a throw of 3' at a rate of 0.5 Hz. Four receivers could be operated at the same time. The $J = 3-2$ rotational lines of $\text{H}^{12}\text{C}^{14}\text{N}$ (hereafter referred to as HCN) at 265.9 GHz, H^{13}CN (259.0 GHz) and HC^{15}N (258.2 GHz) were measured (Table 1). The beam diameter (half-power beam width) of 9.5'' corresponded to 11300 km at the distance of the comet. Observations were undertaken in good atmospheric conditions (3–5 mm precipitable water). The lines were observed both at low (1 or 2 MHz) and high (62 kHz) spectral resolutions. Spectra are shown in Figs 1–2, and line areas are given in Table 1. Observations of HCN, H^{13}CN and HC^{15}N were not entirely simultaneous (Table 1). However, several strong lines (CS $J = 3-2$, CH_3CN $J = 7-6$ (147 GHz) and CH_3OH $J = 3-2$ (145 GHz) lines) were continuously observed to monitor the comet activity. Their intensities decreased by 12% during the period (5.5 hours long) when HCN, H^{13}CN and HC^{15}N data were acquired. This variation was taken into account when deriving the isotopic ratios.

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TABLE 1
CHARACTERISTICS OF RADIO LINES AND MOLECULAR PRODUCTION RATES IN COMET 17P/HOLMES.

Molecule	Line	Frequency (GHz)	Date UT (October 2007)	Int. time (min)	Line area (K km s ⁻¹)	Opacity	Column density (10 ¹² mol cm ⁻²)	Production rate (10 ²⁶ mol s ⁻¹)
HCN	$J = 3-2$	265.886434	27.96–27.98	15	18.23 ± 0.14	0.56	20.8 ± 0.17	19.85 ± 0.16
HCN	$J = 3-2 F = 2-2$	265.888516	27.96–27.98	15	0.958 ± 0.083	0.03	20.7 ± 1.8	19.74 ± 1.71
HCN	$J = 3-2 F = 3-3$	265.884883	27.96–27.98	15	1.097 ± 0.073	0.03	23.7 ± 1.6	22.60 ± 1.50
H ¹³ CN	$J = 3-2$	259.011798	28.08–28.19	90	0.220 ± 0.037	0.008	1.74 ± 0.30	0.164 ± 0.028
HC ¹⁵ N	$J = 3-2$	258.156996	27.96–28.19	145	0.191 ± 0.021	0.006	1.51 ± 0.16	0.140 ± 0.015

NOTE. — The integration time is on+off source; error bars are 1σ ; line areas refer to the main beam brightness temperature scale; column densities and production rates were determined using the radiative transfer model of Biver et al. (1999) with $x_{ne} = 0.5$ and a steady state coma with $T_{kin} = 45$ K as described in the text. The production rate determined from the whole HCN line (resp. HCN hyperfine components, H¹³CN and HC¹⁵N lines) is 6% lower (resp. 6% higher), using $T_{kin} = 65$ K derived by Dello Russo et al. (2008) from HCN infrared observations on October 27.6, 2007.

In contrast to the HC¹⁵N and H¹³CN lines, the HCN $J = 3-2$ is optically thick (Table 1). Therefore, radiative transfer modeling is required to retrieve the HCN/H¹³CN, HCN/HC¹⁵N isotopic ratios if the HCN $J = 3-2$ line is used in the analysis. The coma of comet Holmes was in a non-equilibrium regime following its outburst, with gas phase species released by icy grains. Unavoidable simplifying model assumptions on the coma structure could introduce systematic opacity-dependent errors in the retrievals and affect the determination of the isotopic ratios. However, the HCN $J = 3-2$ line is split into six hyperfine components, two of which ($F = 2-2$, $F = 3-3$) are well separated from the core of the line, and detected in the HCN spectrum (Fig. 2, Table 1). These hyperfine components have intrinsic line strengths of 3.7% the total strength according to hyperfine statistical weights, and are optically thin (Table 1). HCN/H¹³CN, HCN/HC¹⁵N abundance ratios determined using the HCN optically thin hyperfine components do not depend upon model assumptions on coma temperature, structure and temporal variability, because in this case emission lines from molecules in the same excitation state and in the same regions of the coma are compared. They are directly given by the line intensity ratios, albeit minor corrections accounting for slightly different line frequencies. Using the HCN hyperfine components, and correcting for the non-simultaneity of the HCN, H¹³CN and HC¹⁵N measurements, we derive H¹²CN/H¹³CN = 114 ± 26 and HC¹⁴N/HC¹⁵N = 139 ± 26 .

Column densities and production rates determined with the radiative transfer model of Biver et al. (1999) are given in Table 1. We assumed a steady-state isotropic parent molecule distribution, with a gas velocity of 0.56 km s⁻¹, and a gas kinetic temperature T_{kin} of 45 K inferred from IRAM observations of multiple lines of CH₃OH on October 29.0 UT. Interestingly, similar isotopic ratios are obtained using the HCN production rate deduced from the whole $J = 3-2$ line as with the model independent hyperfine lines method. As shown in Fig. 2, when the day and night side HCN velocities are fixed to 0.6 and 0.4 km s⁻¹, the model provides a satisfactory fit to the shape of the HCN line. This suggests that our description of the HCN spatial distribution and excitation is correct in first approximation. This conclusion is supported by the good agreement between the HCN production rate measured on October 27.6 in a smaller ($\sim 1''$) aperture (Dello Russo et al. 2008) and those reported in this work.

3. THE ¹⁴N/¹⁵N RATIO IN CN IN 17P/HOLMES

High-resolution optical observations were performed to measure the ¹⁴N/¹⁵N and ¹²C/¹³C ratios in CN. Spectra of the B²Σ⁺–X²Σ⁺(0,0) CN band at 388 nm were obtained on

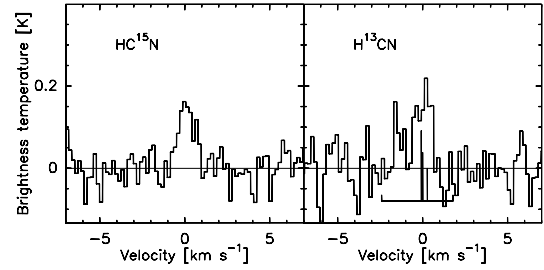


FIG. 1.— Spectra of the $J = 3-2$ lines of HC¹⁵N and H¹³CN in comet 17P/Holmes on 27–28 October 2007. The velocity frame is with respect to the comet rest velocity. The positions and relative intensities of the hyperfine components of H¹³CN $J = 3-2$ are shown.

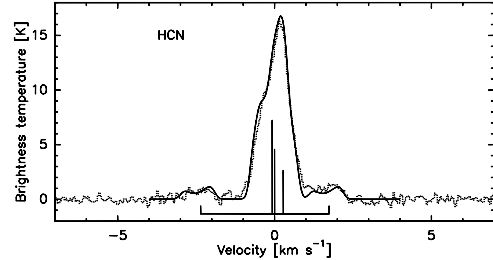


FIG. 2.— Model fitting (continuous line, see text) to the $J = 3-2$ HCN line profile observed in comet 17P/Holmes on Oct. 27.97 UT (dotted line). Positions and relative intensities of the hyperfine components are shown.

October 25.4, 28.3, 29.4, 30.4, 31.4 and November 18.4 and 19.3 2007 UT with the 2DCoudé spectrograph at the 2.7-m Harlan J. Smith telescope of the McDonald Observatory. A series of short exposures, from 30 seconds to 5 minutes, were also collected on October 29.6 2007 UT with the High Resolution Echelle Spectrometer (HIRES) of the Keck 1 telescope installed on Mauna Kea (Hawaii). The observations were carried out under clear weather and low airmass. The slit of the spectrograph was in both cases $\sim 1''$ wide and $\sim 7''$ long providing a resolving power of about $\lambda/\Delta\lambda = 60\,000$ (0.03 Å/pixel). The slit was centered on the false nucleus in the case of the Keck spectra and displaced in the coma (by up to $20''$) for the McDonald exposures (20 min) to reduce the contamination by the strong dust-reflected spectrum. The dust-reflected sunlight underlying the spectral lines (among which are ro-vibrational lines of ¹²C¹⁴N, ¹³C¹⁴N and ¹²C¹⁵N (0,0) band) was removed by subtracting a solar reference spectrum

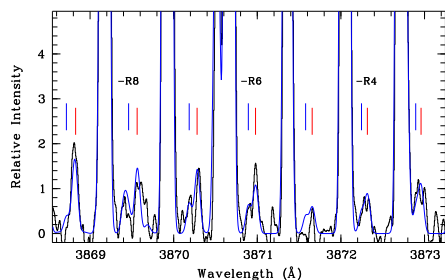


FIG. 3.— A small part of the co-added Keck and McDonald CN (0,0) band spectra of comet 17P/Holmes showing seven R branch lines (R3 to R9) of CN isotopes (black line). The dust-scattered solar spectrum was removed. The positions of the $^{13}\text{C}^{14}\text{N}$ (red ticks) and $^{12}\text{C}^{15}\text{N}$ (blue ticks) lines are indicated; the corresponding lines of the main isotope are on the left. The synthetic spectrum with $^{12}\text{C}/^{13}\text{C}=90$ and $^{14}\text{N}/^{15}\text{N}=165$ is superimposed (blue line).

after the appropriate Doppler shift, profile fitting and normalization were applied (Arpigny et al. 2003; Jehin et al. 2004). The individual CN (0,0) spectra were then combined with an optimal weighting scheme in order to maximize the overall signal-to-noise ratio. Synthetic spectra of $^{12}\text{C}^{14}\text{N}$, $^{13}\text{C}^{14}\text{N}$, $^{12}\text{C}^{15}\text{N}$ were computed for each observing circumstance using a fluorescence model (Zuconi & Festou 1985). We took into account slightly different excitation conditions over the period of the observations caused by small variations of the heliocentric distance and velocity. Collisional effects were empirically estimated by fitting the $^{12}\text{C}^{14}\text{N}$ lines (Manfroid et al. 2005). The synthetic spectra were then co-added in the same way as the data. The isotope mixture was adjusted to best fit the observed continuum-subtracted spectrum. We considered seven R branch lines (R3 to R9), as shown in Fig. 3. The isotopic ratios of $^{12}\text{C}/^{13}\text{C}$ and $^{14}\text{N}/^{15}\text{N}$ in the final co-added Keck and McDonald spectrum are estimated to be 90 ± 20 and 165 ± 40 , respectively.

4. REANALYSIS OF THE COMET HALE-BOPP DATA

The $^{14}\text{N}/^{15}\text{N}$ ratios measured in HCN and CN in comet 17P/Holmes are consistent with each other. In contrast, an $\text{HC}^{14}\text{N}/\text{HC}^{15}\text{N}$ ratio marginally higher than the Earth atmospheric value (272) was reported for comet C/1995 O1 (Hale-Bopp) (Jewitt et al. 1997; Ziurys et al. 1999), to be compared to the value $^{14}\text{N}/^{15}\text{N} = 140 \pm 35$ measured in CN (Arpigny et al. 2003). This led us to reanalyse the measurements made in comet Hale-Bopp.

Our reanalysis of the data of Ziurys et al. (1999) obtained on March 24 and 25, 1997 yields $^{12}\text{C}/^{13}\text{C} = 65 \pm 13$ and $^{14}\text{N}/^{15}\text{N} = 152 \pm 30$ (with a 10% calibration uncertainty included) as compared with the $\text{HC}^{14}\text{N}/\text{HC}^{15}\text{N}$ production rates ratio of 100 ± 20 and 286 ± 82 given by these authors. Ziurys et al. used approximate formulas to analyse the optically thick HCN lines, assuming in addition an inappropriate value for the rotational temperature of HCN. Instead, we used full radiative transfer modeling. Our determinations are likely more reliable as the HCN production rates that we derive from the $J = 1-0$, $2-1$ and $3-2$ HCN lines observed on March 24 are consistent within 5%, while the values inferred by Ziurys et al. from the different lines differ by up to a factor of 2. The HC^{15}N and H^{13}CN $J = 3-2$ lines were observed on March 25. As one cannot exclude day-to-day variations in HCN production from March 24 to 25, the $\text{HC}^{15}\text{N}/\text{H}^{13}\text{CN}$ ratio of 0.43 ± 0.10 , deduced from March 25 data only, could be more secure. In this case, assuming $^{12}\text{C}/^{13}\text{C}$ to be equal to the terrestrial value of 89, we deduce $^{14}\text{N}/^{15}\text{N} = 207 \pm 48$.

The data obtained by Jewitt et al. (1997) at the James Clerk

Maxwell Telescope (JCMT) are public and available from the Canadian Astronomy Data Centre. Reanalysing these data, we found: 1) the HCN data were acquired during night time near sunrise, while the H^{13}CN line and especially the HC^{15}N line were observed later during daytime; the beam efficiency may have degraded by 20% according to JCMT specifications; 2) the HCN line was observed with the receiver B3 tuned in double sideband (DSB), while the other lines were observed in single sideband (SSB); HCN spectra of calibration sources obtained with the same receiver DSB tuning as used for the cometary observations show signals in excess of 15% with respect to reference spectra of the sources; 3) the H^{13}CN $J = 4-3$ line is blended with the SO_2 $13_{2,12}-12_{1,11}$ line at 345.338538 GHz (Lis et al. 1997); using the SO_2/HCN production rate ratio determined in comet Hale-Bopp (Bockelée-Morvan et al. 2000), we estimate that it affects the H^{13}CN line intensity by 20%; 4) more critical, the HC^{15}N spectra present scan-to-scan intensity variations by a factor of 2 (a factor of 10 above the fluctuations related to the statistical noise) that are likely of instrumental origin. Taking these corrections into account, we infer $^{12}\text{C}/^{13}\text{C} = 94 \pm 8$ and $^{14}\text{N}/^{15}\text{N} = 205 \pm 70$, while the values given in Jewitt et al. (1997) are 100 ± 12 and 323 ± 46 , respectively. The large uncertainty in our $^{14}\text{N}/^{15}\text{N}$ determination reflects the dispersion of the HC^{15}N measurements.

We conclude that the $^{14}\text{N}/^{15}\text{N}$ ratio in HCN is rather uncertain for comet Hale-Bopp but is consistent with the value measured in CN.

5. IMPLICATIONS

The $^{12}\text{C}/^{13}\text{C}$ values in HCN and CN are in agreement with the terrestrial value of 89 and previous measurements in comets (e.g., Bockelée-Morvan et al. 2005).

The $^{14}\text{N}/^{15}\text{N}$ ratios measured in HCN and CN in comet 17P/Holmes both correspond to a factor of two ^{15}N enrichment relative to the Earth atmospheric value, and are consistent with the $^{14}\text{N}/^{15}\text{N}$ ratios measured in CN in a dozen comets which cluster at 141 ± 29 (Arpigny et al. 2003; Hutsemékers et al. 2005; Manfroid et al. 2005; Jehin et al. 2004). The discrepancy between HCN and CN isotopic ratios previously found for comet Hale-Bopp led to the suggestion of a CN production mechanism other than HCN photolysis in cometary atmospheres, possibly from the thermal degradation of ^{15}N -rich refractory organics present in dust grains. This interpretation was supported by the presence of CN jets, by the radial distribution of CN, found to be generally less extended than expected from HCN photodissociation, and by the CN/HCN production rate ratio which exceeds unity in some comets (Fray et al. 2005). However, questions arose of how to explain the equally low $\text{C}^{14}\text{N}/\text{C}^{15}\text{N}$ value observed in comet Hale-Bopp at large heliocentric distance where the CN radicals were expected to be mainly HCN photodissociation products (Manfroid et al. 2005; Rauer et al. 2003). Our reanalysis of the Hale-Bopp data, which shows that the $^{14}\text{N}/^{15}\text{N}$ ratio in HCN encompasses the CN value, solves this issue. Also, a similar isotopic ratio in HCN and CN provides a better explanation to the uniform values of the $\text{C}^{14}\text{N}/\text{C}^{15}\text{N}$ ratio among comets which exhibit large differences in dust-to-gas production rate ratios.

Our isotopic measurements are compatible with HCN being the prime parent of CN in cometary atmospheres. For comet Hale-Bopp, the production rates of the two species were found approximately equal (Rauer et al. 2003; Fray et al. 2005). The complex and variable structure of 17P/Holmes's

coma after its outburst makes comparisons of the HCN and CN production rates difficult for this comet. However, Dello Russo et al. (2008) note that the good agreement between the HCN/H₂O abundance determined from infrared spectra and the OH/CN abundance ratio measured with narrowband photometry (Schleicher 2007) is consistent with CN being mainly produced by HCN photolysis. Yet, we cannot exclude that, in other comets, CN has other major progenitors (dust or gas-phase species) sharing the same low ¹⁴N/¹⁵N isotopic ratio, which would mean that ¹⁵N enrichment is possibly a general property of CN-bearing compounds in comets.

Comets Hale-Bopp and 17P/Holmes are of different dynamical families, the former originating from the Oort cloud, and the latter from the trans-Neptunian scattered disc. CN anomalous nitrogen isotopic composition is observed in a number of comets of these two dynamical populations. Hence, comets issued from these two reservoirs likely exhibit similar anomalous N isotopic composition in HCN.

The ¹⁵N excess measured in cometary HCN ($\delta^{15}\text{N} \sim 1000$ per mil relative to the Earth atmospheric value) and CN daughter-product cannot result from isotopic fractionation in the comet atmosphere. It is comparable to the extreme enrichments measured in interplanetary dust particles (Floss et al. 2006) (IDPs) and carbonaceous meteorites (Busemann et al. 2006). High $\delta^{15}\text{N}$ values are also present at the submicrometre scale in the dust particles collected by the Stardust mission in comet 81P/Wild 2 (McKeegan et al. 2006). In IDPs and meteorites, the ¹⁵N-rich nitrogen is carried by non-volatile macromolecular organic material and is generally believed to be a remnant of interstellar chemistry, though a nucleosynthesis source is often considered as an alternative mechanism given the presence of presolar grains with high ¹⁵N excesses in the matrix of meteorites (e.g., Zinner 1998). These complex organics possibly formed from UV or cosmic radiation processing of simple ices in the presolar cloud or, at later stages, in the cold regions of the solar nebula. The enrichments reported here provide the first evidence for the presence of high ¹⁵N anomalies in the volatiles that composed the icy phase of the outer solar nebula, possibly representing the precursors of the primitive refractory organics.

Cometary HCN and H₂O ices exhibit strong enrichments in deuterium with respect to the cosmic D/H value (Ehrenfreund et al. 2005; Bockelée-Morvan et al. 2005), which are believed to reflect ion-molecule and gas-

grain fractionation reactions that took place at low temperature in the early phases of Solar System formation or in the natal molecular cloud. The interpretation of the ¹⁵N enrichment in HCN (by a factor of 3 with respect to the protosolar value in the main nitrogen reservoir; Fouchet et al. 2004; Meibom et al. 2007) is not as compelling as for deuterium, because there is still little evidence for N isotopic fractionation in the interstellar medium (Ikeda et al. 2002) and predicted ¹⁵N enhancements in HCN for exchange reactions involving the main nitrogen reservoir N₂ are modest (Terzieva et al. 2000). Rodgers & Charnley (2004, 2008) show that highly fractionated NH₃ ice could have formed in interstellar or protosolar material if N₂ was converted into atomic nitrogen. While this mechanism is attractive to account for the high ¹⁵N excesses in primitive refractory organics, if synthesized from NH₃, the chemical network for explaining the HCN isotopic anomaly in comets still has to be proposed. Alternative mechanisms include a nucleosynthetic origin and photochemical self-shielding in the solar nebula, similar to that proposed for explaining the oxygen isotope anomalies in meteorites (Clayton 2002a,b). Irrespective of the mechanism, protosolar HCN never isotopically equilibrated with nebular gas at later phases of Solar System evolution.

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